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13. ABSTRACT (Maximum 200 words)
We have performed experimental and theoretical investigations of novel coherent effects in semiconductors, including excitonic Rabi oscillations, soliton-like pulse propagation at the exciton resonance, the nonlinear optical skin effect, interaction-induced polarization rotation in uniaxially strained quantum wells, light-induced ultrafast heavy-to-light hole population transfer, and the coherently coupled heavy-hole-light hole Stark effect. We have identified the role of semiconductor-specific aspects and contrasted the investigated effects with their two- and three—level counterparts. In general, we found semiconductor-specific aspects, such as the bandstructure and the Coulomb interaction, to modify these effects significantly.

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NOVEL COHERENT EFFECTS IN SEMICONDUCTORS

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August 1, 1995 through June 30, 2000

N. PEYGHAMBARIAN, PRINCIPAL INVESTIGATOR
R. BINDER, CO-PRINCIPAL INVESTIGATOR

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UNIVERSITY OF ARIZONA
OPTICAL SCIENCES CENTER
TUCSON, AZ 85721

ARO Final Report
(Time period covert: 8/1/1995 – 6/31/2000)

1. CO-PRINCIPAL INVESTIGATORS

Nasser Peyghambarian (Principal Investigator)
Professor
Optical Sciences Center
University of Arizona

Rudolf H. Binder (Co-Principal Investigator)
Associate Professor
Optical Sciences Center
University of Arizona

2. STATEMENT OF THE PROBLEMS STUDIED

During the time period covered by the ARO grant, we have investigated several coherent nonlinear effects in semiconductors. The investigations include both experimental and theoretical studies. They include various geometries (bulk semiconductors, semiconductor quantum wells, semiconductor micro cavities and vertical-cavity surface-emitting lasers) and a number of different optical excitation conditions. Common to all investigations is the issue of the ultrafast, coherent, nonlinear optical response of the semiconductor in the spectral vicinity of the fundamental band gap (especially the heavy-hole-exciton and light-hole-exciton response). Our studies included light-pulse propagation dynamics, vectorial polarization dynamics, and polarization-dependent excitonic population dynamics. The central issue common to all those individual studies was the role of semiconductor-specific many-body effects; in other words, the question of whether the observed and/or predicted effects can be understood in terms simple 2-level or 3-level dynamics. Quite generally, the answer to this question determines how future novel ultrafast optical semiconductor devices can be optimized in their performance.

Basically, we found in practically all areas significant deviations of the observed and/or predicted effects from simple 2-level or 3-level dynamics. The following is a list of individual aspects of this investigation and the main results obtained either experimentally, or theoretically, or through a combined theory-experiment analysis.

3. SUMMARY OF THE EXPERIMENTAL RESULTS

1. We performed and reported an experimental study on coherent high intensity pulse propagation on the free exciton resonance in CdSe. Similar to the effect of self-induced transparency in atomic vapors our observations include pulse reshaping, pulse break-up, high transmission rates, and a minimum in the velocity dispersion for pulses tuned to the vicinity of the free exciton resonance. Self-induced transparency is a coherent pulse propagation phenomenon characterized by the almost unattenuated propagation of a pulse of 2π area through several Beer's law absorption length. The observation of this effect requires the pulse duration to be smaller than the dephasing time of the relevant material excitation, otherwise incoherent pulse propagation occurs that is determined by linear absorption and, possibly, incoherent nonlinear absorption effects. Our system of choice are epitaxially grown CdSe crystals that exhibit an exciton resonance well separated from other bound and unbound electron-hole pair excitations with optical

thickness α L of 1.7 and 7, respectively. For the thinner sample the pulse transmission shows polariton beating as supported by model calculations. In the transmission of transform limited 180 fs pulses through the thicker sample we observe a temporal pulse break-up for intensities between 2 and 6 GW/cm^2 that provides a clear indication for coherent long distance propagation. At even higher intensities self-phase modulation causes characteristic temporal and spectral pulse break-up. Our studies show that coherent, low loss propagation of short optical pulses very close to an exciton absorption resonance in a semiconductor is indeed possible.

2. A theoretical study of soliton-like pulse propagation has been performed for both semiconductors and, as an additional investigation, for conventional nonlinear materials. In the case of semiconductors, we have found that certain preciously known analytical solutions for solitons in conventional materials can also describe the soliton-like solution in semiconductors, although the underlying physics in both cases are completely different. Based on the one-dimensional propagation model of Maxwell's equation in the slow-varying envelope approximation, and the two-band semiconductor Bloch equations restricted to single-exciton inter-band polarizations, the temporal soliton-like character of moderate-intensity picosecond light pulses has been found to be dominated by excitonic exchange interactions. In contrast, phase-space blocking effects have been found to play only a minor role. Detailed comparisons with the phenomenon of self-induced transparency (SIT) have revealed that the important McCall-Hahn area theorem, well known for 2-level atoms, takes a completely different form in semiconductors. Whereas in 2-level systems the pulse area of the stable solutions is independent of the pulse duration, there is a distinct non-trivial relation between these two pulse characteristics in semiconductors.

3. We verified experimentally the dynamic nonlinear optical skin effect in a semiconductor. This effect leads to an internal reflection at a propagating boundary between areas of high and low exciton densities. To provide experimental proof of this theoretically predicted nonlinear optical effect, we studied in detail the properties of reflected light upon the incidence of intense femtosecond laser pulses onto a ZnSe crystal that is characterized by high optical nonlinearity at the fundamental exciton resonance. In a single beam experiment the dynamic nonlinear optical skin effect is manifested by an increasing amount of light that is reflected at photon energies extending to more than 20 meV below the energy of the exciton absorption resonance. The reflectivity far away from the exciton transition is unusually high and we obtained reflectivity values that are even larger than unity at photon energies four times the FWHM below the exciton resonance. A vivid explanation is that the frequency of the photons reflected at the (propagating) internal boundary between areas of high and low exciton densities is red-shifted due to the 'moving mirror' (Doppler)effect. In a second step, this frequency shift of the internally reflected light was used to study the propagation of the density front. Performing pump-probe reflection experiments with 150 fs pulses we showed that the Doppler-shifted reflection is present only for the duration of the pump pulse, indicating that the front propagation decelerates directly after the pump pulse leaves the sample. The experimental results agree very well with advanced model calculations that model the density front excitation and propagation deceleration within approximately 100 fs. We find that the density front excited by the strong fs laser pulse can propagate with a velocity of up to 10^6 m/s.

4. The following new coherent optical effect in semiconductors has been predicted: interaction –induced polarization rotation in uni-axially strained quantum wells. More precisely, we predict that high-intensity circularly-polarized ultra-short light pulses which are reflected at a uni-axially strained quantum well will induce a strong and time dependent change of ellipticity of the reflected light. This is caused by intrinsic exciton-exciton interactions. The theory on which these results are based is known as Hartree-Fock multi-band semiconductor Bloch theory.

5. In addition to basic ultrafast nonlinear optical effects in semiconductors, we have investigated vectorial polarization properties of VCSELs. First, we derived and classified the vectorial eigenmodes of VCSELs.

The core of the theory is a generalized vectorial transfer matrix method, and the basic ingredients are the cylindrical hybrid mode solutions of Maxwell's equations known for optical fibers. Combining the fiber mode theory with the generalized transfer matrix approach yields a complete characterization of optical mode frequencies, cavity-losses, longitudinal and transverse amplitude patterns of the electric and magnetic field in the VCSEL cavity, as well as the vector polarization characteristics of the fields. Furthermore, we have performed an investigation of the vectorial stability of linearly polarized fundamental modes in VCSELs with small amounts of anisotropic strain or stress in the optically active semiconductor quantum well. Our theory is a microscopic theory that includes the mutual dependence of dichroism and birefringence, and, based on this theory, we showed that for given nonzero strain one expects three pump current regimes, out of which two correspond to one stable linear polarization and one that corresponds to a bistable regime.

6. As a theoretical extension and application of the theory of dark states in semi-conductors, we have studied and predicted the light-induced adiabatic population transfer in semiconductor quantum wells. The population transfer under consideration involves heavy-hole and light-hole bands in p-doped semiconductor quantum wells. The investigation was based on an appropriate generalization of the multi-band semiconductor Bloch equations. In particular, the influence of many-body effects due to the Coulomb interaction between charge carriers in semiconductors has been studied. Based on our theory we predicted that this type of hole-population transfer is indeed possible, and we found that dynamic energy renormalizations due to Coulomb exchange interactions as well as quasi-thermalization of charge carriers significantly modifies the transfer dynamics as compared to that in simple 3-level systems.

7. Investigating the coherent response of a semiconductor quantum well to a strong resonant light field we reported the first experimental observation of multiple excitonic Rabi oscillations in a semiconductor material. Optical Rabi oscillations are amongst the most fundamental examples of coherent nonlinear light-matter interactions. In atomic and molecular two-level systems, optical Rabi oscillations are well established. Exposed to a strong stationary light field, the electron population oscillates between the lower and upper states with the Rabi frequency that is proportional to the dipole moment and the light field. Experimental difficulties arise from the fact that coherent processes in semiconductors are confined to much shorter times since the phases of the semiconductor excitations randomize over a characteristic time of hundreds of femtoseconds. Thus, experiments must be done with subpicosecond light pulses. Our experimental observation of excitonic Rabi oscillations in semiconductors was based on a unique pump-probe scheme. We excited the Rabi oscillations with a relatively long (770 fs) pump pulse and probed with a relatively short (150 fs) probe pulse to time gate the density oscillations. Utilizing pump and probe pulses that can be independently tuned in wavelength and temporally shaped we were able to create Rabi oscillations in the heavy-hole exciton density of an InGaAs quantum well and probe the transmission changes at the light-hole exciton transition. Since the light-hole exciton and the heavy-hole exciton both derive from the same conduction band oscillations in the heavy-hole exciton density manifest themselves in oscillations of the probe pulse absorption at the light-hole exciton resonance. We were also able to determine the dependence of the Rabi frequency upon pulse intensity. We found that the change in Rabi frequency with light intensity follows a square root behavior as in atomic two-level systems. However, modeling the experiment with the semiconductor Bloch equations shows that the Rabi frequency is renormalized due to Comlomb effects. Our results on this fundamental effect allowed us to analyze in a quantitative way analogies and principal differences in the coherent nonlinear response between two-level atoms and excitons in semiconductors.

8. Another extension of the theory of dark states and, more generally, 3-band dynamics is the area of 3-band Rabi oscillations. This is another novel coherent effect that we predicted theoretically. In contrast to 2-level and 2-band Rabi oscillations, 3-level and 3-band Rabi oscillations have been found to offer a rich variety of oscillation dynamics involving both, radiative and non-radiative (Raman) transitions. We have

analyzed excitonic 3-band Rabi oscillations in semiconductor quantum wells and found that the combination of band-coupling effects and Coulomb effects drastically modifies the 3-band Rabi oscillations as compared to those in simple 3-level systems.

9. We investigated experimentally and theoretically the coherently coupled optical Stark effect in a semiconductor three-state system under off-resonant excitation. This issue is a logical extension of the study of coherent exciton dynamics under resonant excitation condition (see above) that became possible by the granted six-month extension of the proposal. Semiconductor quantum wells that exhibit pronounced and well-separated heavy hole and light hole exciton resonances are in certain respects analogous to a three-state V-system. The optical response at the two resonances is coupled through a shared conduction band, as exhibited, for example, in our previous measurement of multiple excitonic optical Rabi oscillations. Here, we report on the observation of coherently coupled optical Stark shifts of heavy hole and light hole exciton resonances in an InGaAs multiple quantum well. The shifts are induced by 1.2 ps pulses pumping below the heavy hole exciton without excitation of a real exciton or carrier population. Since the splitting between heavy and light hole exciton resonance is even larger than the heavy hole exciton to pump photon energy detuning direct excitation of virtual light hole excitons is largely prohibit. This scheme enables us to study the heavy and light hole exciton dynamics solely induced by coherently driven virtual heavy hole excitons. We measured differential absorption spectra from weak, broadband probe pulses that reveal the dynamics of the semiconductor response at both heavy hole and light hole exciton resonance simultaneously. By employing both counter-circularly-polarized and co-circularly-polarized pump and probe, we were able to correlate shifts of heavy and light hole exciton states that share electron states with the same spin. In doing so, we have made the first measurement of the coherent coupling in a semiconductor three-band system under off-resonant excitation. The theoretical analysis shows that the leading contribution to the coherently coupled light hole exciton shift results from the shared electron state. Higher order Coulomb correlations play a minor rule as long as the pump energy is more than the biexciton binding energy below the heavy hole exciton resonance.

4. PERSONNEL SUPPORTED

Dr. Rudolf H. Binder, Assistant Professor
Dr. Axel Schülzgen, Assistant Research Professor
Dr. Kai Wundke, Postdoctoral Research Associate
Dr. Dariusz Burak, Postdoctoral Researcher
Ilya Ruminantsev, Graduate Student
Zhenghan Yang, Graduate Student
Jason Auxier, PhD Student
Oliver Hennig, MS, 1998
Michael Donovan, PhD, scheduled for Oct. 2000
George Mahs, PhD 1996

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